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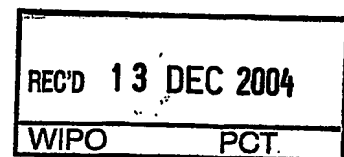
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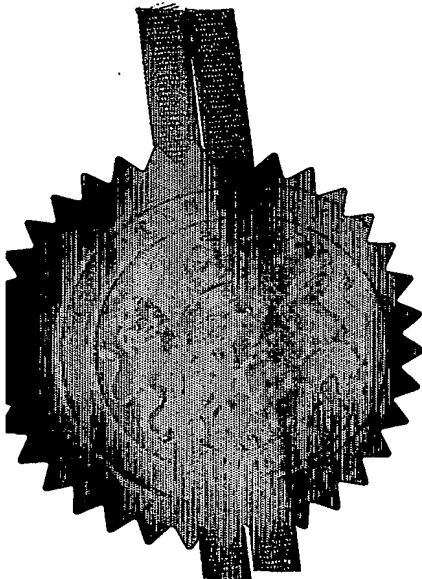


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SMC 80629/GB/P1

2. Patent application number

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0326980.0

3. Full name, address and postcode of the or of each applicant (underline all surnames)

Avecia Limited
Hexagon House
Blackley
Manchester, M9 8ZS
United Kingdom
07764137001 ✓

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

GB

4. Title of the invention

COMPOUNDS

5. Name of your agent (if you have one)

MAYALL, John

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

Avecia Limited
Hexagon House
Blackley
Manchester, M9 8ZS
United Kingdom

Patents ADP number (if you know it)

0244515802 07764137001

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Country	Priority application number (if you know it)	Date of filing (day / month / year)
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7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application	Date of filing (day / month / year)
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8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

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- b) there is an inventor who is not named as an applicant, or
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Patents Form 1/77

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Continuation sheets of this form

Description

12

Claim(s)

01

Abstract

Drawing(s)

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Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 2/77)

Request for substantive examination (Patents Form 10/77)

Any other documents (please specify)

11.

I/We request the grant of a patent on the basis of this application.

Signature

G. Shepherd

Date 20/11/2003

Avecia Limited Authorised Signatory

12. Name and daytime telephone number of person to contact in the United Kingdom

Mrs K.M. Pinder/Mrs G. Shepherd 0161 721 1361/2

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Patents Form 1/77

COMPOUNDS

This invention relates to compounds suitable for use as dyes, to inks and to their use in ink jet printing ("IJP"). IJP is a non-impact printing technique in which droplets of ink are ejected through a fine nozzle onto a substrate without bringing the nozzle into contact with the substrate.

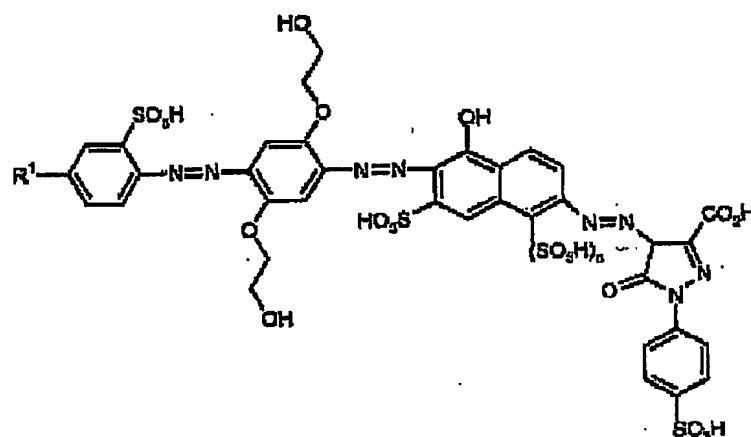
There are many demanding performance requirements for dyes and inks used in IJP. For example they desirably provide sharp, non-feathered images having good water-fastness, ozone-fastness, light-fastness and optical density. The inks are often required to dry quickly when applied to a substrate to prevent smudging, but they should not form a crust over the tip of an ink jet nozzle because this will stop the printer from working. The inks should also be stable to storage over time without decomposing or forming a precipitate which could block the fine nozzle.

JP10195320 describes dyes including tris-azo dyes carrying a pyrazolyl azo group and their use in the coloration of paper and pulp.

US patent application 2001/0027734 describes metal complexes of tris-azo dyes derived from tris-azo molecules optionally containing a (substituted) pyrazolylazo moiety. The copper complexes are said to be particularly preferred.

We have surprisingly found that certain un-metallised compounds provide valuable colorants for ink jet printing inks.

According to the present invention there is provided a compound of Formula (1) or salt thereof:



Formula (1)

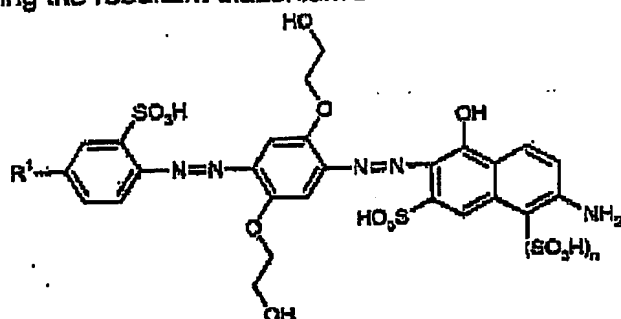
wherein:

R¹ is C₁₋₄alkyl or C₁₋₄alkoxy; and

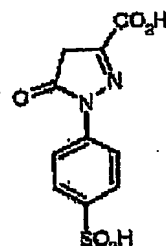
n is 0 or 1.

R¹ is preferably methyl or methoxy.

The compounds of Formula (1) may be prepared by diazotising a compound of the Formula (2), wherein n and R¹ are as hereinbefore defined, to give a diazonium salt and coupling the resultant diazonium salt with a compound of Formula (3):



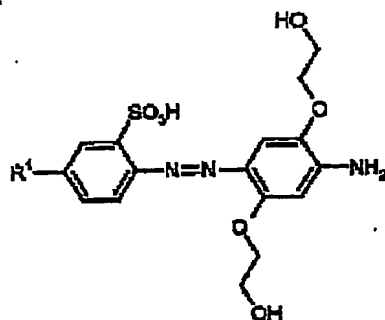
Formula (2)



Formula (3).

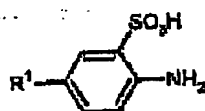
The diazotisation is preferably performed at a temperature of 0°C to 10°C. Preferably the diazotisation is performed in water, preferably at a pH below 7. Dilute mineral acid, e.g. HCl or H₂SO₄, may be used to achieve the desired pH conditions.

The compound of Formula (2) may be prepared by diazotising a compound of Formula (4) to give a diazonium salt and coupling the resultant diazonium salt with 1-hydroxy-3-sulpho-7-amino naphthylene optionally carrying a sulpho group at the 5-position, wherein R¹ is as hereinbefore defined:



Formula (4).

The compound of Formula (4) may be prepared by diazotising a compound of Formula (5) to give a diazonium salt and coupling the resultant diazonium salt with 2,5-bis-(2-acetoxyethoxy)aniline, followed by hydrolysis of the acetoxy groups, wherein R¹ is as hereinbefore defined:



Formula (5).

Reaction conditions for the above processes are those generally used in the dyestuff art, for example as described in for example EP 0356080.

When the compounds of Formula (1) are in the form of a salt the preferred salts are alkali metal salts (especially lithium, sodium and potassium salts), ammonium and substituted ammonium salts and mixtures thereof. Especially preferred salts are sodium, potassium and lithium salts, salts with ammonia and volatile amines and mixtures thereof. The lithium salts have good solubility, forming particularly storage stable inks with low toxicity and low tendency to block ink jet nozzles.

The compounds of Formula (1) may be converted into a desired salt using known techniques. For example, an alkali metal salt of a compound of Formula (1) may be converted into the ammonium or substituted ammonia salt by dissolving an alkali metal salt of the compound in water, acidifying with a mineral acid and adjusting the pH of the solution to pH 9 to 9.5 with ammonia or the amine and removing the alkali metal cations by dialysis or by use of an ion exchange resin.

Examples of amines which may be used to form such salts include methylamine, dimethylamine, trimethylamine, ethylamine, n-propylamine, iso-propylamine, n-butylamine, iso-butylamine, sec-butylamine, tert-butylamine, piperidine, pyridine, morpholine, allylamine, diethylamine, triethylamine, tetramethyl amine and mixtures thereof. It is not essential that the compounds of Formula (1) are completely in the form of the ammonium salt or substituted ammonium salt and mixed alkali metal and either ammonium salt or substituted ammonium salt are effective, especially those in which at least 50% of the cations are ammonium or substituted ammonium ions.

Still further salts are those with the counter ions described in US 5830265, claim 1, integer (b), which are included herein by reference thereto.

The compounds of Formula (1) may exist in tautomeric forms other than those shown in this specification. These tautomers are included within the scope of the present claims.

A second aspect of the invention concerns a composition comprising a compound of Formula (1) as described above and a liquid medium, wherein the liquid medium comprises water and an organic solvent.

A preferred composition according to the second aspect of the invention comprises:

- (a) from 0.01 to 80 parts of a compound of Formula (1) or salt thereof; and
- (b) from 70 to 99.99 parts of a liquid medium;

wherein the liquid medium comprises an organic solvent and all parts are by weight and the number of parts of (a)+(b)=100.

The number of parts of component (a) is preferably from 0.1 to 20, more preferably from 0.5 to 15, and especially from 1 to 5 parts. The number of parts of component (b) is preferably from 99.9 to 80, more preferably from 99.5 to 85, especially from 99 to 95 parts.

Preferably component (a) is completely dissolved in component (b). Preferably component (a) has a solubility in component (b) at 20°C of at least 10%. This allows the preparation of liquid dye concentrates which may be used to prepare inks and reduces the chance of the dye precipitating if evaporation of the liquid medium occurs during storage.

5 Preferred liquid media comprise water and an organic solvent, preferably in a weight ratio of water to organic solvent of 99:1 to 1:99, more preferably 99:1 to 50:50 and especially 95:5 to 50:20.

10 It is preferred that the organic solvent is a water-miscible organic solvent or a mixture of such solvents. Preferred water-miscible organic solvents include C₁₋₆-alkanols, preferably methanol, ethanol, n-propanol, isopropanol, n-butanol, sec-butanol, tert-butanol, n-pentanol, cyclopentanol and cyclohexanol; linear amides, preferably dimethylformamide or dimethylacetamide; ketones and ketone-alcohols, preferably acetone, methyl ether ketone, cyclohexanone and diacetone alcohol; water-miscible ethers, preferably tetrahydrofuran and dioxane; diols, preferably diols having from 2 to 12 carbon atoms, for example
15 pentane-1,5-diol, ethylene glycol, propylene glycol, butylene glycol, pentyleneglycol, hexyleneglycol and thiodiglycol and oligo- and poly-alkyleneglycols, preferably diethylene glycol, triethylene glycol, polyethylene glycol and polypropylene glycol; triols, preferably glycerol and 1,2,6-hexanetriol; mono-C₁₋₄-alkyl ethers of diols, preferably mono-C₁₋₄-alkyl ethers of diols having 2 to 12 carbon atoms, especially 2-methoxyethanol, 2-(2-methoxyethoxy)ethanol, 2-(2-ethoxyethoxy)ethanol, 2-[2-(2-methoxyethoxy)ethoxy]ethanol, 2-[2-(2-ethoxyethoxy)ethoxy]ethanol and ethyleneglycol monoallylether; cyclic amides, preferably 2-pyrrolidone, N-methyl-2-pyrrolidone, N-ethyl-2-pyrrolidone, caprolactam and 1,3-dimethylimidazolidone; cyclic esters, preferably caprolactone; sulfoxides, preferably dimethyl sulphoxide and sulpholane. Preferably the liquid medium comprises water and 2
20 or more, especially from 2 to 8, water-soluble organic solvents.

25 Especially preferred water-soluble organic solvents are cyclic amides, especially 2-pyrrolidone, N-methyl-pyrrolidone and N-ethyl-pyrrolidone; diols, especially 1,5-pentane diol, ethyleneglycol, thiodiglycol, diethyleneglycol and triethyleneglycol; and mono- C₁₋₄-alkyl and C₁₋₄-alkyl ethers of diols, more preferably mono- C₁₋₄-alkyl ethers of diols having 2 to 12 carbon atoms, especially ((2-methoxy-2)-ethoxy)-2-ethoxyethanol.

30 The compounds of the invention may be used as the sole colorant in inks because of their attractive black shade. However, if desired, one may combine the present compounds with one or more further colorants if a slightly different shade is required for a particular end use. The further colorants are preferably dyes. When further colorants are
35 included in the ink these are preferably selected from black, magenta, cyan and yellow colorants and combinations thereof.

Suitable further black colorants include different colorants of the same invention, C.I. Food Black 2, C.I. Direct Black 19, C.I. Reactive Black 31, PRO-JET™ Fast Black 2, C.I. Direct Black 195; C.I. Direct Black 168; and black dyes described in patents by

Lexmark (e.g. EP 0 639,178 A2, Example 1, 2, 3, 4 and 5), Orient Chemicals (e.g. EP 0 347 803 A2, pages 5-6, azo dyes 3, 4, 5, 6, 7, 8, 12, 13, 14, 15 and 16) and Seiko Epson Corporation.

Suitable further magenta colorants include PRO-JET™ Fast Magenta 2.

Suitable further cyan colorants include phthalocyanine colorants, C.I. Direct Blue 199 and C.I. Acid Blue 99.

Suitable further yellow colorants include C.I. Direct Yellow 142; C.I. Direct Yellow 132; C.I. Direct Yellow 86; C.I. Direct Yellow 85; C.I. Direct Yellow 173; and C.I. Acid Yellow 23.

The ink may also contain additional components conventionally used in ink jet printing inks, for example viscosity and surface tension modifiers, corrosion inhibitors, biocides, fogging reducing additives and surfactants which may be ionic or non-ionic.

The pH of the composition is preferably from 4 to 11, more preferably from 7 to 10.

The viscosity of the composition at 25°C is preferably less than 50cP, more preferably less than 20 cP and especially less than 5cP.

When the compositions according to the second aspect of the invention are used as ink jet printing compositions, the composition preferably has a concentration of halide ions of less than 500 parts per million, more preferably less than 100 parts per million. It is especially preferred that the composition has less than 100, more preferably less than 50 parts per million of divalent and trivalent metals, wherein parts refer to parts by weight relative to the total weight of the composition. We have found that purifying the compositions to reduce the concentration of these undesirable ions reduces nozzle blockage in ink jet printing heads, particularly in thermal ink jet printers. A further aspect of the invention provides a process for printing an image on a substrate comprising applying thereto a composition according to the second aspect of the present invention to the substrate by means of an ink jet printer.

The ink jet printer preferably applies the composition to the substrate in the form of droplets which are ejected through a small orifice onto the substrate. Preferred ink jet printers are piezoelectric ink jet printers and thermal ink jet printers. In thermal ink jet printers, programmed pulses of heat are applied to the composition in a reservoir by means of a resistor adjacent to the orifice, thereby causing the composition to be ejected in the form of small droplets directed towards the substrate during relative movement between the substrate and the orifice. In piezoelectric ink jet printers the oscillation of a small crystal causes ejection of the composition from the orifice.

The substrate is preferably paper, plastic, a textile, metal or glass, more preferably a treated substrate such as a coated paper or coated plastic, especially coated paper. Preferred plain or treated papers are papers which may have an acid, alkaline or neutral character. Examples of commercially-available treated papers include HP Premium Coated Paper, HP Photopaper (both available from Hewlett Packard Inc.); Stylus Pro 720

dpi Coated Paper, Epson Photo Quality Glossy Film, Epson Photo Quality Glossy Paper (all available from Seiko Epson Corp.); Canon HR 101 High Resolution Paper, Canon GP 201 Glossy Paper, Canon HG 101 and HG201 High Gloss Film and Canon PR101 (all available from Canon).

A further aspect of the present invention provides a paper, an overhead projector slide or a textile material printed with a composition, a compound or by means of a process as hereinbefore defined.

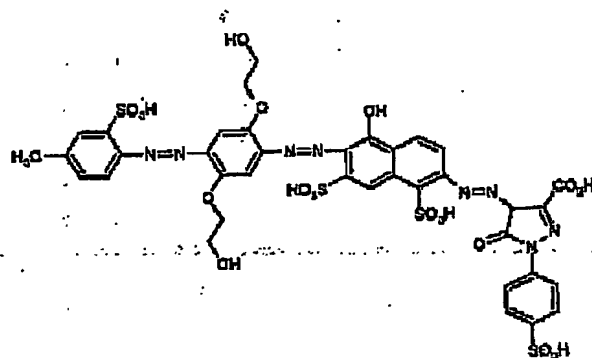
A still further aspect of the present invention provides an ink jet printer cartridge, optionally refillable, comprising one or more chambers and a composition, wherein the composition is present in at least one of the chambers and the composition is as defined in the second aspect of the present invention.

The present compounds and compositions provide prints of attractive, neutral black shades which are particularly well suited for the ink jet printing of text and images. The compositions have good storage stability and low tendency to block the very fine nozzles used in ink jet printers. Furthermore, the resultant images have good optical density, light-fastness, wet-fastness and resistance to fading in the presence of oxidising air pollutants.

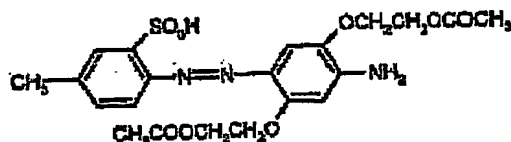
The invention is further illustrated by the following Examples in which all parts and percentages are by weight unless specified otherwise.

Example 1

Preparation of:



Step 1: Preparation of

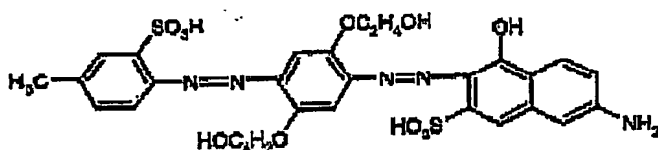


4-Aminotoluene-3-sulphonic acid (28.05g) dissolved in water (500mls) and the pH adjusted to 7-8 by addition of lithium hydroxide (2M). Sodium nitrite (11.4g) was added to the solution and the mixture then added to a mixture of ice and water to which

concentrated hydrochloric acid (45 ml) had been added. The reaction was further stirred for 1 hour at 0-10°C. Excess nitrous acid was then removed by the addition of sulphamic acid to give a diazonium salt.

2,5-Bis-(2-acetoxyethoxy)aniline (59.4g) was dissolved in acetone (600 ml) and the solution added with stirring to the above prepared diazonium salt. The coupling mixture was then stirred overnight at room temperature. The precipitated product was isolated by filtration and washed with water (2 litres). The product was then stirred in acetone (2 litres), filtered and washed with little acetone before drying at 50°C. Yield = 66.8g.

Step 2: Preparation of:



The monoazo compound prepared as described in Step 1 above (23.95g, Mol.In 479) was suspended in N-methylpyrrolidone (250ml) and sodium nitrite (7g) was added. After stirring for 15 minutes at room temperature a complete solution was obtained. To the stirred solution was added dilute hydrochloric acid (15ml conc HCl in 100ml with water) over 5 minutes (exothermic) and stirring was continued for 2 hours allowing to self cool to room temperature. Excess nitrous acid was removed by the addition of sulphamic acid.

6-amino-1-naphthol-3-sulphonic acid (14g) was dissolved in water (200ml) adjusting to pH 10 by addition of lithium hydroxide (2M). The solution was stirred and cooled to 0-10°C and the diazonium salt solution was slowly added, maintaining the pH between 10-10.5 by addition of lithium hydroxide (2M) as necessary. The mixture was further stirred for 1 hour at 0-10°C before allowing to warm to room temperature. Lithium hydroxide (10g) was added and stirring was continued at room temperature until thin layer chromatography showed hydrolysis was complete. Lithium chloride solution (40%w/v) was then slowly added. The product was isolated by filtration at 70°C and washed with lithium chloride solution (50%w/v, 500 ml). The filtrate was suspended in water (500 ml) and the suspension drowned into acetone (5 litres). The acetone was removed by decantation and water added to obtain thick suspension. The pH of the suspension adjusted to 1 using dilute hydrochloric acid before drowning into acetone (5 litres), isolating by filtration and drying at 70°C. Yield = 38.7g

Step 3: Preparation of title compound:

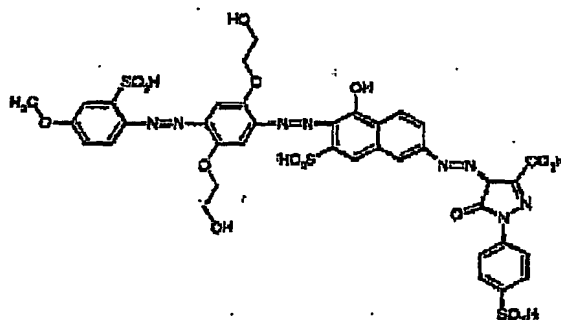
The product of step 2 above (37g, Mol.In 1028) was suspended in water (1 litre) and the pH adjusted to 7-8 with addition of lithium hydroxide solution (2M). N-

methylpyrrolidone (100 ml) was added to obtain a complete solution. Sodium nitrite (2.8g) was added to the solution together with Calsolene™ oil (1g) (Calsolene is a trademark of ICI plc). The mixture was then added to a stirred mixture of ice/water containing concentrated hydrochloric acid (15ml) and further stirred for about 2 hours at 0-10°C. Excess nitrous acid was then removed by the addition of sulphamic acid.

1-(4-Sulphophenyl)-3-carboxypyrazol-5-one (12g) was dissolved in water (100ml) and the pH adjusted to 7-8 by addition of lithium hydroxide (2M). The solution was then added to above stirred diazonium salt solution, the pH adjusted to 6-7 by addition of lithium hydroxide (2M), and stirred for 1 hour at 0-10°C. The solution was drowned into acetone (5litres) and the precipitated product isolated by filtration and washed with acetone. The product was dissolved in water (500ml), dialysed to low conductivity before screening through a 0.45µ nylon filter and isolation by evaporation to dryness at 70°C. Yield = 29.5g.

Example 2

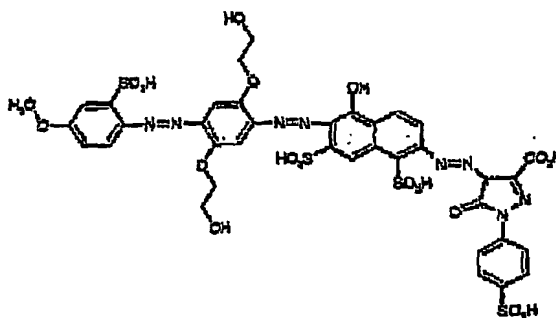
Preparation of:



The method of Example 1 was repeated except that in Step 1, 2-amino-5-methoxybenzenesulphonic acid (30.46g) was used in place of 4-amino-3-toluenesulphonic acid.

Example 3

Preparation of:



The method of Example 1 was repeated except that in Step 1, 2-amino-5-methoxybenzenesulphonic acid (30.45g) was used in place of 4-amino-3-toluenesulphonic acid and in Step 2, 6-amino-1-naphthol-3,5-disulphonic acid (16.8g) was used in place of 6-amino-1-naphthol-3-sulphonic acid.

Example 4 - Inks

Inks may be prepared containing the compounds from Example 1 to Example 3 according to the following formulation:

2-Pyrrolidone	5 parts
Thiodiglycol	5 parts
Surfynol™ 4651	part
Dye	3 parts
Water	amount required to make up to 100 parts

Surfynol™ 465 is a surfactant available from Air Products and Chemicals Inc., USA.

Further inks may be prepared according to Tables I and II wherein the compound described in the first column is the compound made in the above example of the same number. Numbers quoted in the second column onwards refer to the number of parts of the relevant ingredient and all parts are by weight. The inks may be applied to paper by thermal or piezo ink jet printing.

The following abbreviations are used in Tables I and II:

PG = propylene glycol
DEG = diethylene glycol
NMP = N-methyl pyrrolidone
DMK = dimethylketone
IPA = isopropanol
MEOH = methanol
2P = 2-pyrrolidone
MIBK = methylisobutyl ketone
P12 = propane-1,2-diol
BDL = butane-2,3-diol
CET = cetyl ammonium bromide
PHO = Na_2HPO_4 and
TBT = tertiary butanol
TDG = thiodiglycol

Example 5 – Ink Jet Printing

The inks prepared in Example 4 may be incorporated into an empty cartridge of a Hewlett Packard DeskJet 550C™ ink jet printer and printed onto Xerox X Acid™ paper, or onto photopapers such as PR101 (Canon) or SEC PM (Epson), to give prints of excellent shade, optical density, light and ozone fastness.

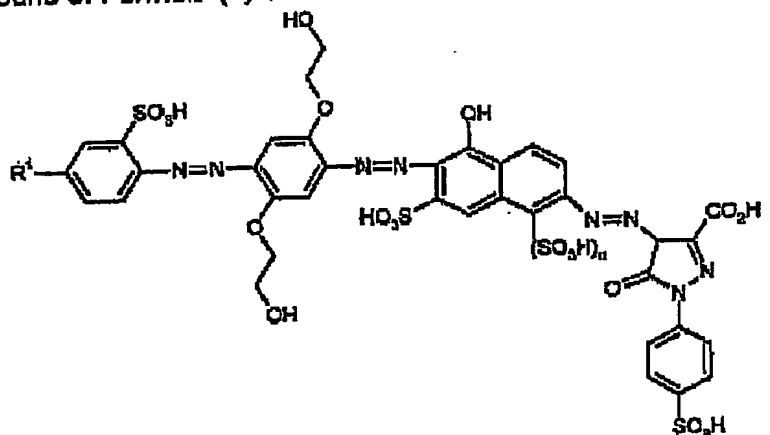
5

TABLE II

Dye	Dye Content	Water	PG	DEG	NMP	CET	TBT	TDG	BDL	PHO	2P	P12
1	3.0	80	15			0.2				1.2	5	5
2	9.0	90		5								
3	1.5	85	5	5		0.15	5.0	0.2		0.12		
1	2.5	90		6	4							
2	3.1	82	4	8		0.3			5	0.2		6
3	0.9	85		10								
1	8.0	90		5	5			0.3				
2	4.0	70		10	4				1		4	11
3	2.2	75	4	10	3				2		6	
1	10.0	91			6						3	
2	9.0	78		9	7		3.0			0.95	5	
3	5.0	78	5	11							6	
1	5.4	86			7						7	
2	2.1	70	5	5	5	0.1	0.2	0.1	5	0.1	5	
3	2.0	90		10								
1	2	88										
2	5	78			5			10			5	
3	8	70	2		8			12			5	
1	10	80						15			12	
2	10	80		10				8				

CLAIMS

1. A compound of Formula (1) or salt thereof:



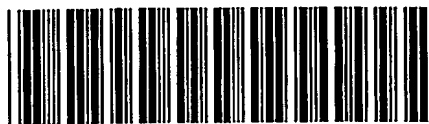
Formula (1)

wherein:

R^1 is C_{1-4} alkyl or C_{1-4} alkoxy; and
 n is 0 or 1.

2. A compound according to claim 1 wherein R^1 is methyl or methoxy.
3. A composition which comprises a compound according to claim 1 or claim 2 and a liquid medium, wherein the liquid medium comprises water and an organic solvent.
4. A composition according to claim 3 which contains a further colorant selected from black, magenta, cyan or yellow colorants.
5. A process for printing an image on a substrate comprising applying thereto a composition according to claim 3 or 4 by means of an ink jet printer.
6. A paper, an overhead projector slide or a textile material printed with a compound according to claim 1 or 2, a composition according to claim 3 or 4, or by means of a process according to claim 5.
7. An ink jet printer cartridge, optionally refillable, comprising one or more chambers and a composition, wherein the composition is present in at least one of the chambers and the composition is as defined in claims 3 or 4.

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